

**Claims:**

1. A process for preparing an organic-inorganic hybrid film material, which comprises the steps of:
  - (a) reacting an aromatic diamine with aromatic dianhydride at a temperature of from room temperature to 50°C to give poly(amic acid), in which an equivalent ratio of the aromatic diamine to the aromatic dianhydride is less than 2;
  - (b) coupling the poly(amic acid) from step (a) with an amino coupling agent having a general formula of  $\text{NH}_2\text{-R}^1\text{-Si}(\text{R}^2)_3$  in which  $\text{R}^1$  is a  $\text{C}_{1-6}$  alkylene or phenylene group,  $\text{R}^2$ 's are the same or different and represent  $\text{C}_{1-6}$  alkoxy group, to give a poly(amic acid) terminated with the amino coupling agent, in which the equivalent of the added coupling agent is less than that of the diamine;
  - (c) subjecting a monomer of formula  $\text{R}^3\text{-Si}(\text{R}^4)_3$  (wherein  $\text{R}^3$  represents a hydrogen,  $\text{C}_{1-6}$  alkyl,  $\text{C}_{2-6}$  alkenyl, and phenyl, and  $\text{R}^4$ 's are the same or different and represent a halogen,  $\text{C}_{1-6}$  alkoxy,  $\text{C}_{2-6}$  alkenyloxy, and phenoxy group) to sol-gel reaction in the presence of acidic catalyst in a solvent at a temperature of from room temperature to 100°C, to give poly(silsesquioxane); wherein the acidic catalyst is added in an amount sufficient to maintain a pH of the reaction mixture at a range from 1 to 4;
  - (d) hydrolyzing the poly(amic acid) terminated with the amino coupling agent from step (b) in the presence of deionized water and then coupling with the poly(silsesquioxane) from step (c), to give a slurry of poly(amic acid)-poly(silsesquioxane) composite material; wherein the amount of deionized water for hydrolyzing the amino coupling agent which is coupled to the poly(amic acid) is molar equivalent to or slight excess the moles of terminal alkoxy group present in the poly(amic acid) terminated with the amino coupling agent;
  - (e) applying the resultant composite material slurry on a substrate, curing the coated

slurry at an elevated temperature to produce an organic-inorganic hybrid film material of polyimide/poly(silsesquioxane).

2. The process according to claim 1, which further comprises a step of distillating the poly(silsesquioxane) at reduced pressure to remove byproduct methanol after step (c).

3. The process according to claim 1, wherein said aromatic dianhydride is selected from the group consisting of pyromellitic dianhydride (PMDA), 4,4'-bipthalic dianhydride (BPDA), 4,4'-hexa-fluoroisopropylidene-diphthalic dianhydride (6FDA), 1-(trifluoromethyl)-2,3,5,6-benzenetetracarboxylic dianhydride (P3FDA), 1,4-di(trifluoromethyl)-2,3,5,6-benzenetetracarboxylic dianhydride (P6GDA), 1-(3',4'-dicarboxy-phenyl)-1,3,3-tri-methyl-indan-5,6-dicarboxylic dianhydride, 1-(3',4'-dicarboxy-phenyl)-1,3,3-trimethyl-indan-6,7-dicarboxylic dianhydride, 1-(3',4'-dicarboxy-phenyl)-3-methyl-indan-5,6-dicarboxylic dianhydride, 1-(3',4'-dicarboxy-phenyl)-3-methyl-indan-6,7-dicarboxylic dianhydride, 2,3,9,10-perylene-tetracarboxylic dianhydride, 1,4,5,8-naphthalene-tetracarboxylic dianhydride, 2,6-dichloro-naphthalene-1,4,5,8-tetracarboxylic dianhydride, 2,7-dichloro-naphthalene-1,4,5,8-tetracarboxylic dianhydride, 2,3,6,7-tetrachloro-naphthalene-2,4,5,8-tetracarboxylic dianhydride, phenanthrene-1,8,9,10-tetracarboxylic dianhydride, 3,3',4'4'-benzophenone-tetracarboxylic dianhydride, 2,2',3,3'-benzophenone-tetracarboxylic dianhydride, 3,3',4',4'-biphenyl-tetracarboxylic dianhydride, 2,2',3,3'-biphenyl-tetracarboxylic dianhydride, 4,4'-isopropylidene-diphthalic anhydride, 3,3'-isopropylidene-diphthalic anhydride, 4,4'-oxy-diphthalic anhydride, 4,4'-sulfonyl-diphthalic anhydride, 3,3'-oxy-diphthalic anhydride, 4,4'-methylene-diphthalic anhydride, 4,4'-thio-diphthalic anhydride, 4,4'-ethylidene-diphthalic anhydride, 2,3,6,7-naphthalene-tetracarboxylic dianhydride, 1,2,4,5-naphthalene-tetracarboxylic dianhydride, 1,2,5,6-naphthalene-tetracarboxylic

dianhydride, benzene-1,2,3,4-tetracarboxylic dianhydride, pyrazine-2,3,5,6-tetracarboxylic dianhydride, and a combination thereof.

4. The process according to claim 1, wherein said aromatic diamine is selected from the group consisting of 4,4'-oxy-dianiline (ODA), 5-amino-1-(4'-aminophenyl)-1,3,3-trimethyl-indane; 6-amino-1-(4'-aminophenyl)-1,3,3-trimethyl-indane, 4,4'-methylene-bis(o-chloro-aniline), 3,3'-dichloro-dibenzidme, 3,3'-sulfonyl-dianiline, 4,4'-diamino-benzophenone, 1,5 -diamino-naphthalene, bis(4-aminophenyl)diethyl silane, bis(4-aminophenyl)diphenyl silane, bis(4-aminophenyl)ethyl phosphine oxide, N-[bis(4-aminophenyl)]-N-methyl amine, N-(bis(4-aminophenyl))N-phenyl amine, 4,4'-methylene-bis(2-methyl-aniline), 4,4'-methylene-bis(2-methoxy-aniline), 5,5'-methylene-bis(2-aminophenol), 4,4'-methylene-bis(2-methyl-aniline), 4,4'-oxy-bis(2-methoxy-aniline), 4,4'-oxy-bis(2-cliloro-aniline), 2,2'-bis(4-aminophenol), 5,5'-oxy-bis(2-aminophenol), 4,4-thio-bis(2-methyl-aniime), 4,4'-thio-bis(2-methoxy-aniline), 4,4'-thio-bis(2-chloro-aniline), 4,4'-sulfonyl-bis(2-methyl-aniline), 4,4'-sulfonyl-bis(2-ethoxy-aniline), 4,4'-sulfonyl-bis(2-chloro-aniline), 5,5'-sulfonyl-bis(2-aminophenol), 3,3'-dimethyl-4,4'-diamino-benzophenone, 3,3'-dimethoxy-4,4'-diamino-benzophenone, 3,3'-dichloro-4,4'-diamino-benzophenone, 4,4'-diamino-biphenyl, m-phenylenediamine, p-phenylene-diamine, 4,4'-methylene-dianiline, 4,4'-thio-dianiline, 4,4'-sulfonyl-dianiline, 4,4'-isopropylidene-dianiline, 3,3'-dimethyl-dibenzidine, 3,3'-dimethoxy-dibenzidine, 3,3'-dicarboxy-dibenzidine, 2,4-tolyl-diamine, 2,5-tolyl-diamine, 2,6-tolyl-diamine, m-xylyl-diamine, 2,4-diamino-5-chloro-toluene, 2,4-diamino-6-chloro-toluene, and a combination thereof.

5. The process according to claim 1, wherein said amino coupling agent of formula  $\text{NH}_2\text{-R}^1\text{-Si}(\text{R}^2)_3$ , is selected from the group consisting of 3-aminopropyl trimethoxy silane (APrTMS), 3-aminopropyl triethyl silane(APrTES), 3-aminophenyl trimethoxy silane(APTMS), 3-aminophenyl triethoxy silane (APTES), and a

combination thereof.

6. The process according to claim 1, wherein said monomer of formula  $R^3-Si(R^4)_3$  is selected from the group consisting of methyl trimethoxy silane (MTMS), trimethoxy silane (TMS), triethoxy silane (TES), methyl triethoxy silane (MTES), phenyl trimethoxy silane (PTMS), phenyl triethoxy silane (PTES), vinyl trimethoxy silane (VTMS), vinyl triethoxy silane (VTEs), trichlorosilane, methyl trichloro silane, phenyl trichloro silane, vinyl trichloro silane, and a combination thereof.

7. An organic-inorganic hybrid film material produced by the process according to claim 1.

8. A process for preparing an organic-inorganic hybrid film material, which comprises the steps of:

(a1) reacting an aromatic diamine with aromatic dianhydride at a temperature of from room temperature to 50°C to give poly(amic acid), in which an equivalent ratio of the aromatic diamine to the aromatic dianhydride is less than 2;

(b1) coupling the poly(amic acid) from step (a1) with an amino coupling agent having a general formula of  $NH_2-R^1-Si(R^2)_3$ , in which  $R^1$  is a  $C_{1-6}$  alkylene or phenylene group,  $R^2$ s are the same or different and represent  $C_{1-6}$  alkoxy group, to give a poly(amic acid) terminated with the amino coupling agent, in which the equivalent of the added amino coupling agent is less than that of the diamine;

(d1) hydrolyzing the poly(amic acid) terminated with the amino coupling agent from step (b1) in the presence of deionized water and then coupling with silicon alkoxide, to give a slurry of poly(amic acid)-silicon alkoxide composite material; wherein the amount of deionized water for hydrolyzing the amino coupling agent which is coupled to the poly(amic acid) is molar equivalent to or slight excess the moles of terminal alkoxy group present in the poly(amic acid) terminated with the amino coupling agent;

(e1) applying the resultant composite material slurry on a substrate, curing the coated slurry at an elevated temperature to produce an organic-inorganic hybrid film material of polyimide/silicon alkoxide.

9. The process according to claim 8, wherein said aromatic dianhydride is selected from the group consisting of pyromellitic dianhydride (PMDA), 4,4-biphenal dianhydride (BPDA), 4,4'-hexa-fluoroisopropylidene-diphthalic dianhydride (6FDA), 1-(trifluoromethyl)-2,3,5,6-benzenetetracarboxylic dianhydride (P3FDA), 1,4-di(trifluoromethyl)-2,3,5,6-benzenetetracarboxylic dianhydride (P6GDA), 1-(3',4'-dicarboxy-phenyl)-1,3,3-tri-methyl-indan-5,6-dicarboxylic dianhydride, 1-(3',4'-dicarboxy-phenyl)-1,3,3-trimethyl-indan-6,7-dicarboxylic dianhydride, 1-(3',4'-dicarboxy-phenyl)-3-methyl-indan-5,6-dicarboxylic dianhydride, 1-(3',4'-dicarboxy-phenyl)-3-methyl-indan-6,7-dicarboxylic dianhydride, 2,3,9,10-perylene-tetracarboxylic dianhydride, 1,4,5,8-naphthalene-tetracarboxylic dianhydride, 2,6-dichloro-naphthalene-1,4,5,8-tetracarboxylic dianhydride, 2,7-dichloro-naphthalene-1,4,5,8-tetracarboxylic dianhydride, 2,3,6,7-tetrachloro-naphthalene-2,4,5,8-tetracarboxylic dianhydride, phenanthrene-1,8,9,10-tetracarboxylic dianhydride, 3,3',4'4'-benzophenone-tetracarboxylic dianhydride, 2,2',3,3'-benzophenone-tetracarboxylic dianhydride, 3,3',4',4'-biphenyl-tetracarboxylic dianhydride, 2,2',3,3'-biphenyl-tetracarboxylic dianhydride, 4,4'-isopropylidene-diphthalic anhydride, 3,3'-isopropylidene-diphthalic anhydride, 4,4'-oxy-diphthalic anhydride, 4,4'-sulfonyl-diphthalic anhydride, 3,3'-oxy-diphthalic anhydride, 4,4'-methylene-diphthalic anhydride, 4,4'-thio-diphthalic anhydride, 4,4'-ethylidene-diphthalic anhydride, 2,3,6,7-naphthalene-tetracarboxylic dianhydride, 1,2,4,5-naphthalene-tetracarboxylic dianhydride, 1,2,5,6-naphthalene-tetracarboxylic dianhydride, benzene-1,2,3,4-tetracarboxylic dianhydride, pyrazine-2,3,5,6-tetracarboxylic dianhydride, and a combination thereof.

10. The process according to claim 8, wherein said aromatic diamine is selected from the group consisting of 4,4'-oxy-dianiline (ODA), 5-amino-1-(4'-aminophnyl)-1,3,3-trimethyl-indane; 6-amino-1-(4'-aminophnyl)-1,3,3-trimethyl-indane, 4,4'-methylene-bis(o-chloro-aniline), 3,3'-dichloro-dibenzidme, 3,3'-sulfonyl-dianiline, 4,4'-diamino-benzophenone, 1,5 -diamino-naphthalene, bis(4-aminophnyl)diethyl silane, bis(4-aminophnyl)diphenyl silane, bis(4-aminophnyl)ethyl phosphine oxide, N-[bis(4-aminophnyl)]-N-methyl amine, N-(bis(4-aminophnyl))N-phenyl amine, 4,4'-methylene-bis(2-methyl-aniline), 4,4'-methylene-bis(2-methoxy-aniline), 5,5'-methylene-bis(2-aminophenol), 4,4'-methylene-bis(2-methyl-aniline), 4,4'-oxy-bis(2-methoxy-aniline), 4,4'-oxy-bis(2-chloro-aniline), 2,2'-bis(4-aminophenol), 5,5'-oxy-bis(2-aminophenol), 4,4-thio-bis(2-methyl-anime), 4,4'-thio-bis(2-methoxy-aniline), 4,4'-thio-bis(2-chloro-aniline), 4,4'-sulfonyl-bis(2-methyl-aniline), 4,4'-sulfonyl-bis(2-ethoxy-aniline), 4,4'-sulfonyl-bis(2-chloro-aniline), 5,5'-sulfonyl-bis(2-aminophenol), 3,3'-dimethyl-4,4'-diamino-benzophenone, 3,3'-dimethoxy-4,4'-diamino-benzophenone, 3,3'-dichloro-4,4'-diamino-benzophenone, 4,4'-diamino-biphenyl, m-phenylenediamine, p-phenylene-diamine, 4,4'-methylene-dianiline, 4,4'-thio-dianiline, 4,4'-sulfonyl-dianiline, 4,4'-isopropylidene-dianiline, 3,3'-dimethyl-dibenzidine, 3,3'-dimethoxy-dibenzidine, 3,3'-dicarboxy-dibenzidine, 2,4-tolyl-diamine, 2,5-tolyl-diamine, 2,6-tolyl-diamine, m-xylyl-diamine, 2,4-diamino-5-chloro-toluene, 2,4-diamino-6-chloro-toluene, and a combination thereof.

11. The process according to claim 8, wherein said amino coupling agent of formula  $\text{NH}_2\text{-R}^1\text{-Si}(\text{R}^2)_3$  is selected from the group consisting of 3-aminopropyl trimethoxy silane (APrTMS), 3-aminopropyl triethyl silane(APrTES), 3-aminophenyl trimethoxy silane(APTMS), 3-aminophenyl triethoxy silane (APTES), and a combination thereof.

12. The process according to claim 8, wherein said silicon alkoxide is selected

from the group consisting of tetramethoxysilane, tetraethoxysilane, and a combination thereof.

13. An organic-inorganic hybrid film material produced by the process according to claim 8.